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ELECTRICAL RESISTIVITY, MAGNETIC SUSCEPTIBILITY, AND THERMOELECTRIC POWER OF $PtGa_2$

by

L. Hsu,[*] L.W. Zhou,[†] F.L.A. Machado,[†] W.G. Clark[†] and R.S. Williams

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University of California, Los Angeles
Department of Chemistry & Biochemistry and Solid State Sciences Center
Los Angeles, CA 90024-1569

[*]Chemical Engineering, UCLA
[†]Department of Physics, UCLA

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Electrical Resistivity, Magnetic Susceptibility and Thermoelectric Power of $PtGa_2$

Li-Shing Hsu, Lu-Wei Zhou,* F. L. A. Machado,† W. Gilbert Clark

Department of Physics and Solid State Science Center

University of California, Los Angeles, California 90024

and

R. Stanley Williams

Department of Chemistry & Biochemistry and Solid State Science Center

University of California, Los Angeles, California 90024-1569

The electrical resistivity (ρ), magnetic susceptibility (χ) and thermoelectric power (S) of $PtGa_2$ were measured as a function of temperature (T). This compound is metallic at high temperatures, as shown from the room-temperature resistivity value (19$\mu\Omega$-cm) and the linear dependence of the S vs. T curve at temperatures above the Debye temperature (θ_D). It undergoes a superconducting phase transition with a critical temperature (T_c) at zero magnetic field of 2.13K. The density of states (DOS) at the Fermi energy (E_F) at high temperatures obtained from χ and S data are 22% and 15% higher, respectively, than the value obtained previously from a semi-empirical band structure calculation.

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I. INTRODUCTION

At temperatures above 460K, the intermetallic compound $PtGa_2$ forms a pseudobinary system with GaAs, whereas elemental Pt reacts chemically with GaAs to form more stable product compounds.[1] Although $PtGa_2$ is supposed to be a high-temperature phase and only a metastable species at room temperature, it is actually quite robust. Single crystals and thin films of $PtGa_2$ can be grown and examined over the course of years without perceivable disproportionation. Therefore, the study of the electronic and magnetic properties of $PtGa_2$ is important for understanding its behavior as a potential conducting contact or an active component in optoelectronic circuitry.

$PtGa_2$ has the cubic fluorite structure, and is isostructural with AuX_2 (X=Al,Ga,In). Jan and Pearson[2] have reported that $AuGa_2$ is anomalous in the sense that its thermopower is negative at "low" and "high" temperatures while $AuAl_2$ and $AuIn_2$ have positive thermopowers in the temperature range measured (2 to 300K). The ^{71}Ga Knight shift and the magnetic susceptibility of $AuGa_2$ are strongly temperature-dependent in comparison to its Al and In analogues.[3] On the other hand, resistivity[2,4] and specific heat[5] measurements display no anomalous variation with temperature in $AuGa_2$. In Switendick and Narath's nonrelativistic augmented plane wave (APW) band-structure calculation,[6] a flat band ($\Gamma_{2'}$-X_3) lies about 1eV below E_F in $AuGa_2$, while for $AuAl_2$ and $AuIn_2$ this band disperses strongly and crosses E_F. Kim et al.,[7] who included the spin-orbit interaction in their mixed-basis band structure interpolation scheme (MBBSIS) calculation, reproduced this result. It is generally believed that this flat Δ_2 band, derived from Ga 4s-like anti-bonding states, is responsible for the $AuGa_2$ anomalies discussed above. However, the observation that the magnetic susceptibility of $AuGa_2$ between 4.2 and 300K shows a decreasing diamagnetism with decreasing T is still an unresolved issue.[8] In an angle-resolved photoemission spectroscopy (ARPES) study of $AuGa_2$,[9] no peak was observed corresponding to the Δ_2 band, although such a flat band should yield an extremely high density of initial states to be sampled.

The MBBSIS was recently utilized to obtain a semiempirical band structure of $PtGa_2$.[10] The flat Δ_2 band was also present in this semiempirical band structure, because the $AuGa_2$ parameters were used as the starting point in the fit of the DOS to an X-ray photoemission spectrum of the $PtGa_2$ valence band. However, the ^{71}Ga Knight shift of $PtGa_2$ is positive and temperature-independent, and the conductivity exhibits no anomalous behavior between 4.2 and 300K.[11] Since there is no first-principles band-structure calculation and very little experimental data published for $PtGa_2$, the present study was initiated to provide more information about this potentially interesting material. Section II of this paper describes the experimental procedure. In Sec. III, the results are presented and discussed, and Sec. IV concludes this paper.

II. EXPERIMENTAL PROCEDURE

Samples used for the susceptibility measurements were small pieces, with a total weight of 115.4mg, crushed from a $PtGa_2$ single crystal.[12] A Faraday method, utilizing a Cahn balance, was used for the static magnetic susceptibility measurement in a field of 9 kOe. Temperatures from 4.2 to 300K were measured with calibrated carbon-glass and platinum resistors. In order to verify that the observed magnetization was linear in magnetic field, the susceptibility was measured at several field values at room temperature, liquid-nitrogen temperature and liquid-helium temperature. The uncertainty in χ is less than 1%.

For the electrical resistivity and thermoelectric power measurements, the same single crystal was cut with a wire saw into a long slice of roughly $10\times1\times0.5$ mm^3 in size. It was then polished with 5 micron diamond grit and cleaned with acetone just before loading into the dewar. The electrical resistivity was measured with a four-probe method. The thermopower was measured between 4.2 and 300K by establishing a temperature gradient across the sample and measuring the voltage developed against Au leads. The Seebeck coefficient (S) was obtained from the slope of a linear least-squares fit of a series of 30 Seebeck voltage vs. thermal gradient measurements. The absolute Seebeck coefficient was derived after subtracting out the contribution of the Au lead wires from the resultant slope. The uncertainty in S is less than 2.5%.

III. RESULTS AND DISCUSSION

A. Electrical Resistivity

Figure 1 shows the electrical resistivity of $PtGa_2$; there is no anomaly in the ρ vs. T curve, in agreement with the observation reported in Ref. 11. The room temperature resistivity of $PtGa_2$, which is only about eight times larger than that of Au, is compared with those of $AuGa_2$ and Au in Table 1. Compared with the room temperature electrical resistivity values of WSi_2 (35-60 $\mu\Omega$-cm)[14] and $TaSi_2$ (40 $\mu\Omega$-cm)[15], which have been suggested as high temperature non-reactive contacts on GaAs, $PtGa_2$ is a rather good metal and perhaps to be preferred as a contact for devices. However, the residual resistivity ratio ($\rho_{297.5K}/\rho_{4.2K}$) is only 3.34, which indicates that there may be low levels of impurities or vacancies that result from the metastability of the compound.

In a separate set of measurements to be reported elsewhere,[17] the heat capacity and electrical resistivity have been measured from 4.2K down to 0.5K and 1.4K, respectively. Both measurements show a sharp ($\lesssim$ 30 mK wide) superconducting transition at the temperature $T_c = 2.13$K in zero magnetic field. This value for T_c is higher than that reported for $AuGa_2$. A standard analysis of these results gives the values $n(E_F) = 1.56 \pm 0.3$ electrons of both spin directions/eV-unit cell and $\theta_D = 173 \pm 25$K. These values, which are similar to those obtained from the other measurements, are included in Table 1.

B. Magnetic Susceptibility

The measured magnetic susceptibility at 9 kOe is shown in Fig. 2. The small structure shown in χ below 80K will not be considered in the discussion presented here. For $PtGa_2$, χ has two contributions: one is the temperature-independent diamagnetic susceptibility from the Pt- and the Ga-ion core electrons (χ_i^{Pt} and χ_i^{Ga}), and the other is the conduction electron susceptibility (χ_e). The expression for χ_e also has two components: one is the paramagnetic Pauli susceptibility (χ_e^p), and the other is the Landau-Peierls diamagnetic susceptibility (χ_e^d). For noninteracting free electrons at OK, χ_e^p and χ_e^d are given by

$$\chi_e^p = \mu_B^2\, n(E_F) \qquad (1)$$

and

$$\chi_e^d = -\frac{1}{3}\,\mu_B^2 \left(\frac{m_0}{m^*}\right)^2 n(E_F) \quad , \qquad (2)$$

where μ_B is the Bohr magneton, $n(E_F)$ is the DOS at E_F for both spin directions, and m^*/m_0 is the effective mass ratio. The estimated values for χ_i^{Pt} and χ_i^{Ga} are -28 μemu/mole[18] and -9.54 μemu/mole,[19] respectively. The net ionic diamagnetism for $PtGa_2$ is therefore -47.08 μemu/mole. Taking $m^* = m_0$ in Eq. (2) and using χ at 4.2K to represent that at OK, we get $n(E_F)$ =1.40 electrons of both spin directions/eV-unit cell, which is 22% larger than the value calculated by the MBBSIS.[10] In general, electron-electron interactions lead to an enhancement of the Pauli term by a factor $(1-\alpha)^{-1}$, where α is the Stoner enhancement parameter. Using the $n(E_F)$ value from the MBBSIS calculation in the Pauli and the Landau-Peierls terms, we estimate α to be around 0.16. Since α usually lies in the range of 0.1 to 0.5,[20] this means that electron-electron interactions in $PtGa_2$ are very weak. This in turn justifies the use of the free-electron approximation in the above calculation of $n(E_F)$.

C. Thermoelectric Power

The S vs. T curve of $PtGa_2$ is shown in Fig. 3 along with those of Au and of $AuGa_2$. Below 12K, S of $PtGa_2$ becomes negative. This behavior can be attributed to trace magnetic impurity scattering, which has also been observed in Au.[21] The shape of the S vs. T curve of $PtGa_2$ is very similar to that of Au, although the S values of the former are roughly a factor of two larger than those of the latter. This similarity suggests that there are also some similarities in their conduction mechanisms and the topology of their Fermi surfaces. As has already been pointed out,[10] $PtGa_2$ has an Au-like DOS, which explains the gold color of this intermetallic compound.

Since S of $PtGa_2$ remained positive at the highest temperature measured, the electrical conduction is by holes.[22] This behavior is different from that of $AuGa_2$. The calculated flat Δ_2 band of $PtGa_2$, which is located within 0.1eV of E_F in the Γ-X direction in the MBBSIS, may

actually be either above E_F or may disperse more strongly and cross E_F, as in the case of $AuAl_2$ and $AuIn_2$. The Knight shift[11] and magnetic susceptibility data support the latter suggestion, but to be sure about this point, a high-resolution ARPES study of a $PtGa_2$ single crystal should be performed.

The occurrence of a maximum in Fig. 3 for the thermopower of $PtGa_2$ is attributed to the phonon-drag effect.[21] The contribution of this electron-phonon scattering process to S being positive implies a dominance of Umklapp over normal processes. For $PtGa_2$, the temperature of this maximum (T_{max}) is 37.4K, and therefore θ_D is estimated to be $5T_{max}$=187K.[23] The values of θ_D for Au, $AuGa_2$, and $PtGa_2$ determined by various methods are presented in Table 1.

For $T \geq \theta_D$, impurity scattering is negligible compared with thermal scattering and the phonon-drag contribution to S is rather small. Hence, diffusion thermopower (S_d) dominates. For metallic conduction, S_d varies linearly with T,[24] and the free electron expression is: [25]

$$S_d = \left(\frac{\pi^2 k_B^2 \, n(E_F)}{3Ne} \right) T , \qquad (3)$$

where N is the number of electrons per unit cell, e is the electron charge, k_B is the Boltzmann constant, and $n(E_F)$ is the DOS at E_F.

The dashed line in Fig. 3, which has a slope $(1.035 \pm 0.083) \times 10^{-8}$ V/K^2, is the least-squares fit to the data points for T > 187K for $PtGa_2$. This linear dependence of S with T shows that $PtGa_2$ is metallic for $T > \theta_D$. Comparing with Eq.(3) and using N=3, one may determine that $n(E_F)$ = 1.27 electrons of both spin directions/eV-unit cell, which is presented in Table 1 along with those of Au and $AuGa_2$. The N=3 configuration has been used and justified in certain superconducting compounds containing Ga.[26,27] Pauling[28] assigned effective metallic valences of 6 and 3.5 for Pt and Ga, respectively, when they are bonded in intermetallic compounds. The total number of electrons in one unit cell of $PtGa_2$ is 16 (10 from Pt and 3 from each Ga), and,

from simple addition of valence, 13 of them are used to form the Pt-Ga bonds. Therefore, the number of free electrons in one unit cell of $PtGa_2$ is 3. This explains qualitatively the assignment of N=3.

IV. CONCLUSIONS

Transport properties of $PtGa_2$ have been examined to determine the single-electron properties of this metastable intermetallic compound. Above θ_D, the conduction is by holes and is metallic in nature, and at 2.13K there is a sharp superconducting transition. The magnetic susceptibility, thermoelectric power, and specific heat[17] measurements of $PtGa_2$ were all consistent with the one-electron DOS at E_F estimated from the previous MBBSIS calculation,[10] which is thus shown to have useful predictive capabilities.

V. ACKNOWLEDGMENTS

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*Permanent address: Physics Department, Fudan University, Shanghai, China.

†Permanent address: Departamento de Fisica, Universidade Federal de Pernambuco
Recife, Pernambuco BRASIL 50739

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Table I. Comparison of values of ρ, θ_D and $n(E_F)$[a] for Au, $AuGa_2$ and $PtGa_2$.

	ρ($\mu\Omega$-cm) at 20°C	θ_D(K)	$n(E_F)$
Au	2.24[b]	165[c]	0.31[d]
		162.4[e]	0.18[f]
		161.6[g]	0.24[h]
$AuGa_2$	12.9[i]	245[j]	1.12[k]
		235[c]	1.14[m]
		192[m]	
$PtGa_2$	19.05[n]	187[c]	1.09[f]
		173±25[q]	1.27[o]
			1.40[p]
			1.56±0.3[q]

a. DOS at E_F in units of number of electrons per eV per unit cell for both spin directions.

b. Handbook of Chemistry &Physics (College Edition), (Chem Rubber Co.: 1984), p. F-120.

c. Estimated from thermoelectric power measurements, $\theta_D \cong 5\, T_{max}$.

d. Calculated from D. L. Martin's specific heat data (Phys. Rev. **141**, 141 (1966)) using $n(E_F)=3\,\gamma/k_B^2\pi^2$, where γ is the intercept of the C_p/T vs. T curve.

e. From specific heat measurement, D. L. Martin, Phys. Rev. **141**, 141 (1966).

f. From MBBSIS band structure calculation, Ref. 10.

g. From zero-temperature elastic constants measurement, G. A. Alers, in *Physical Acoustics*, W. P. Mason, ed. (Academic Press, Inc., New York), Vol. IIIB, Chap. I.

h. Calculated from thermoelectric power data, Ref. 18, at $T > \theta_D$ using Eq.(3) in this paper and N=1.

i. From resistivity measurement, Ref. 2.

j. Calculated in Ref. 5 from the resisitivity data of Ref. 2.

k. From MBBSIS band structure calculation, Ref. 7.

m. From specific heat measurement, Ref. 5.

n. From resistivity measurement, this work.

o. From thermoelectric power measurement, this work.

p. From magnetic susceptibility measurement, this work.

q. From specific heat measurement, Ref. 17.

FIGURE CAPTIONS

Fig. 1. Electrical resistivity of $PtGa_2$ as a function of temperature from 4.2 to 300K.

Fig. 2. Magnetic susceptibility of $PtGa_2$ and $AuGa_2$ as a function of temperature from 4.2 to 300K. The $AuGa_2$ data were taken from Ref. 3.

Fig. 3. Temperature dependence of the thermoelectric power S(T) for Au, $AuGa_2$, and $PtGa_2$. The data for Au and $AuGa_2$ are taken from Ref. 18 and Ref. 2, respectively. The dashed line is the least-squares fit to $PtGa_2$ data for T>187K.

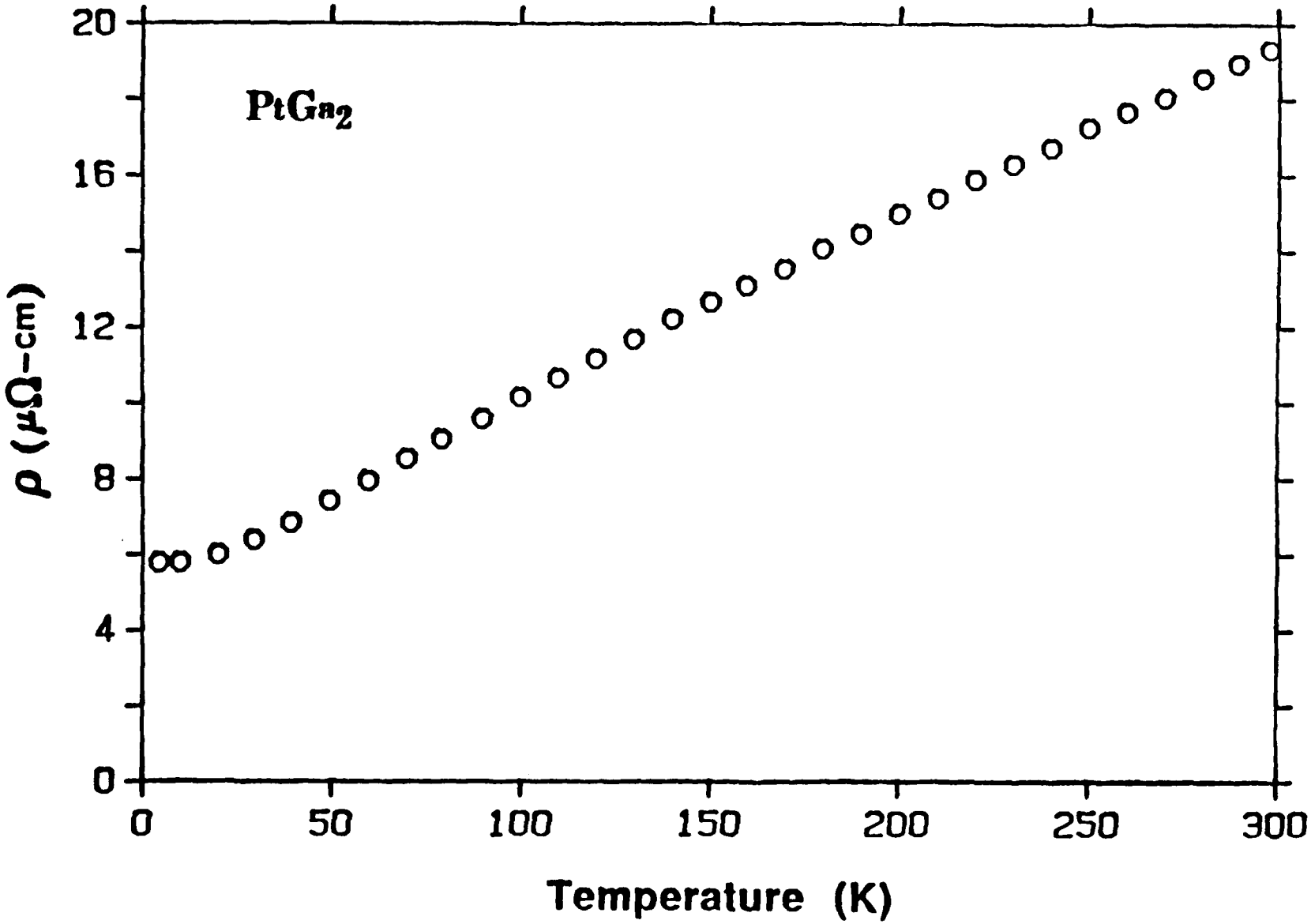
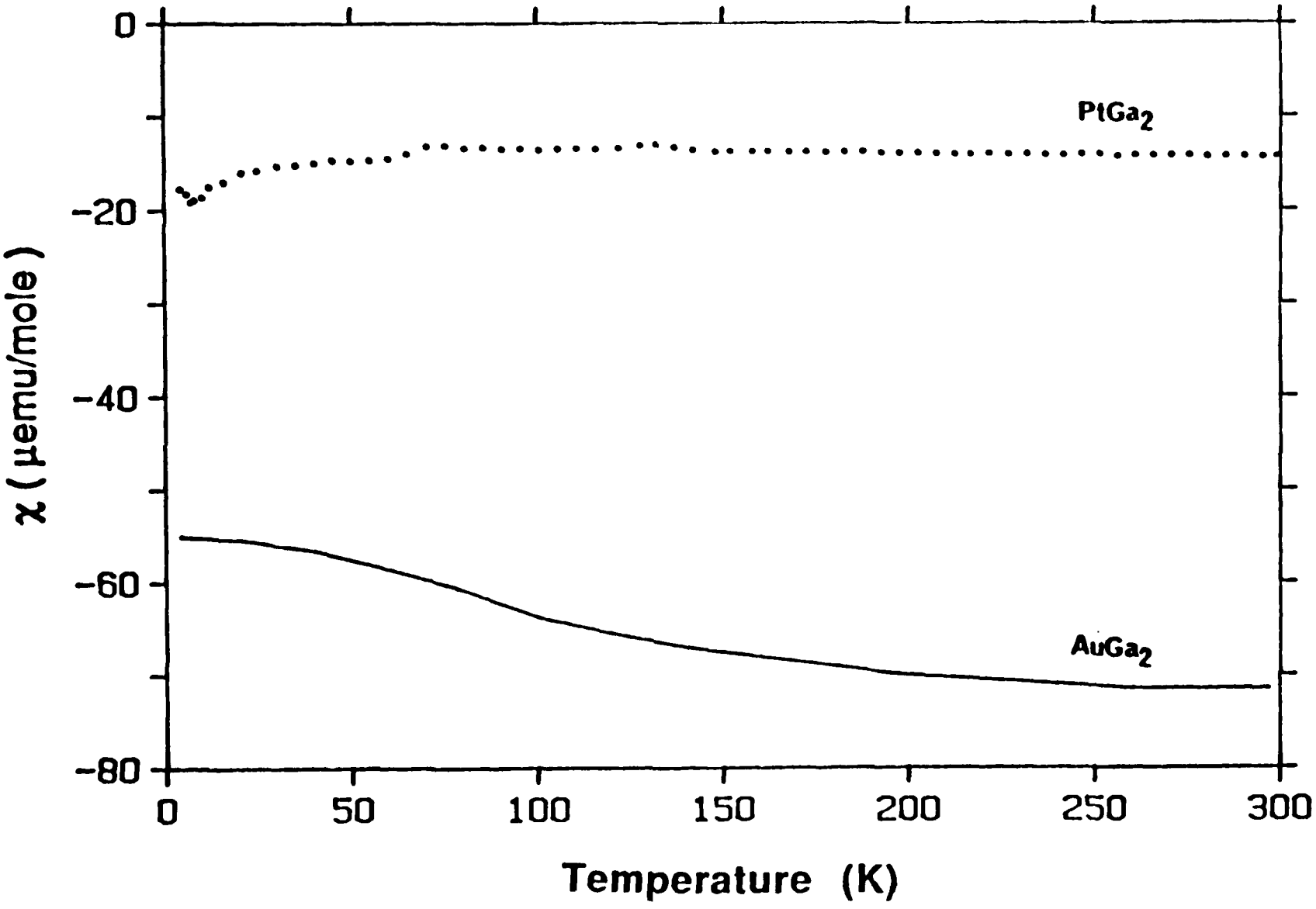


FIG. 1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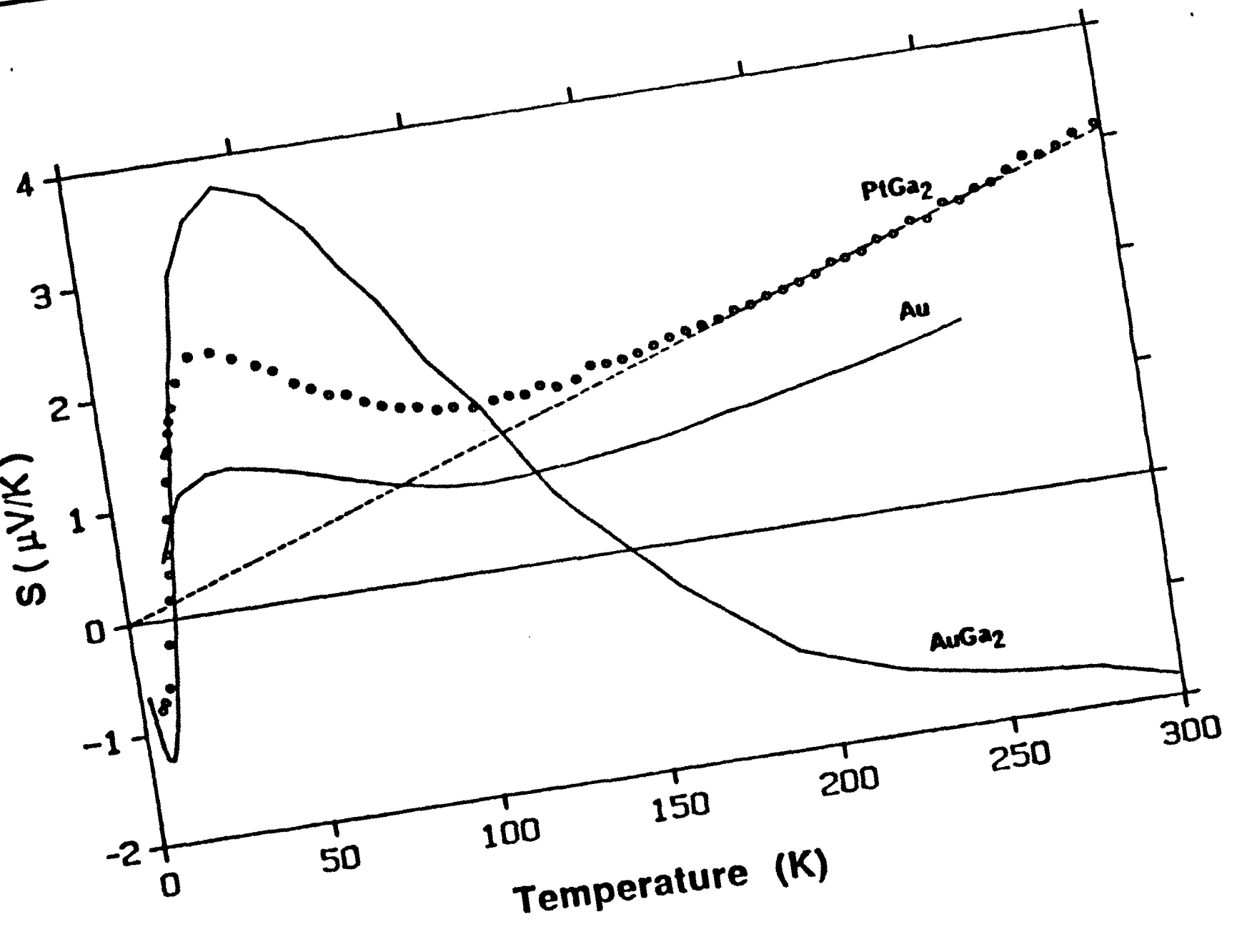
100
150
200
250
300
Temperature (K)

FIG. 2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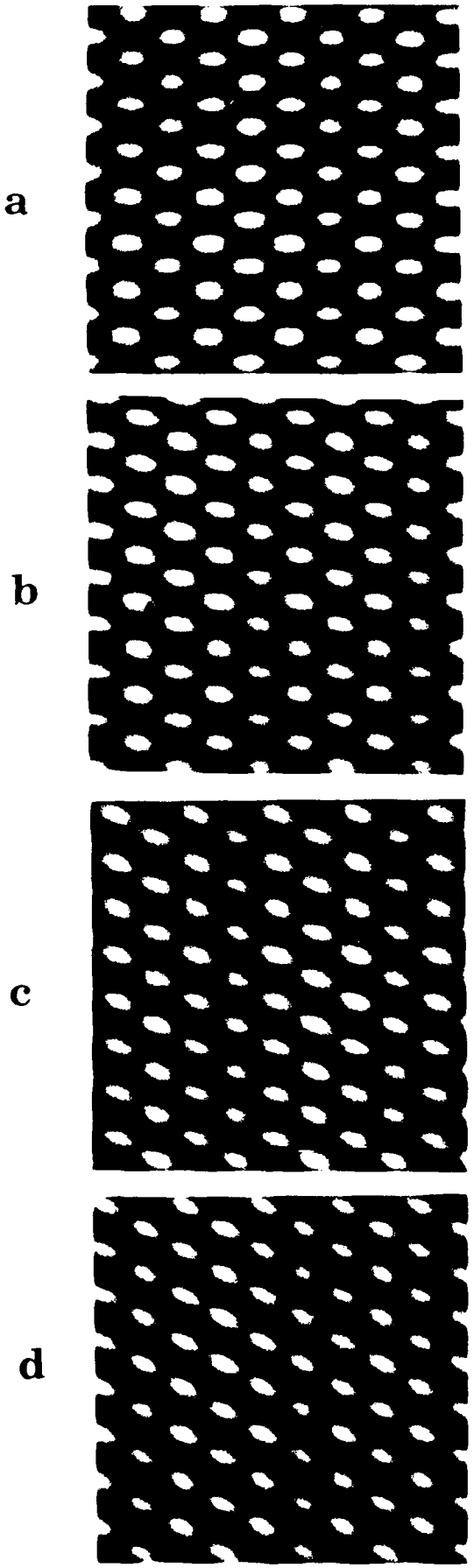
Au
AuGa2

FIG. 3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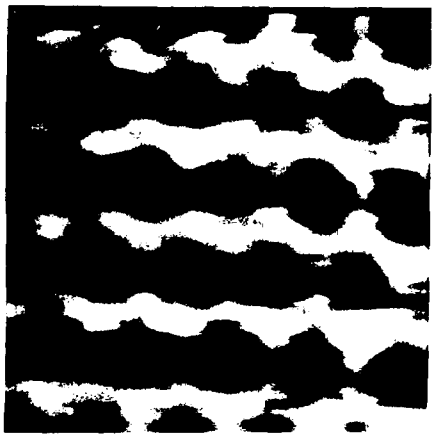

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